STUDY OF PLUTONIUM IN AQUATIC SYSTEMS OF THE ROCKY FLATS ENVIRONS

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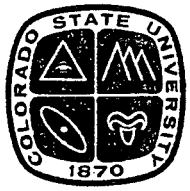

Second Technical Progress Report
Departments of Animal Science
and
Radiology and Radiation Biology

SECOND TECHNICAL PROGRESS REPORT

of the

Department of Animal Sciences
Colorado State University
Fort Collins, Colorado 80521

TO: Dow Chemical Company
Rocky Flats Division
Golden, Colorado 80302

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"The Study of Plutonium in Aquatic Systems of the Rocky Flats Environs"

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I. Introduction

The purposes of this research were to identify and quantify any biological pathways for the movement of Plutonium in the three major water courses at Rocky Flats. The approaches to accomplish these objectives were as follows.

1. Stream and Pond Morphometry data, e.g., flow rate, pond volumes, average and maximum water depths, sediment depths and sediment composition, was collected on a seasonal basis.

2. The inventory of aquatic plants and animals, as well as the terrestrial species in the pond vicinity was continued.

3. Concurrent with the inventory, samples were collected for Plutonium assay. The samples were principally aquatic plants and animals, sediments and water.

4. From results of the previous year of study, laboratory experiments were conducted to elucidate Pu transport mechanism in sediments, terrestrial plants, bacteria and algae.

A substantial effort during the year was spent on revision of the Pu analysis technique. The present analytical method is completely described in this report.

Our sampling protocol also was seriously interrupted during the summer months by pond reconstruction activity. This activity drastically disturbed the pond biology and kinetics but does allow observation of the approach to a new equilibrium in the altered ponds as well as the new one.

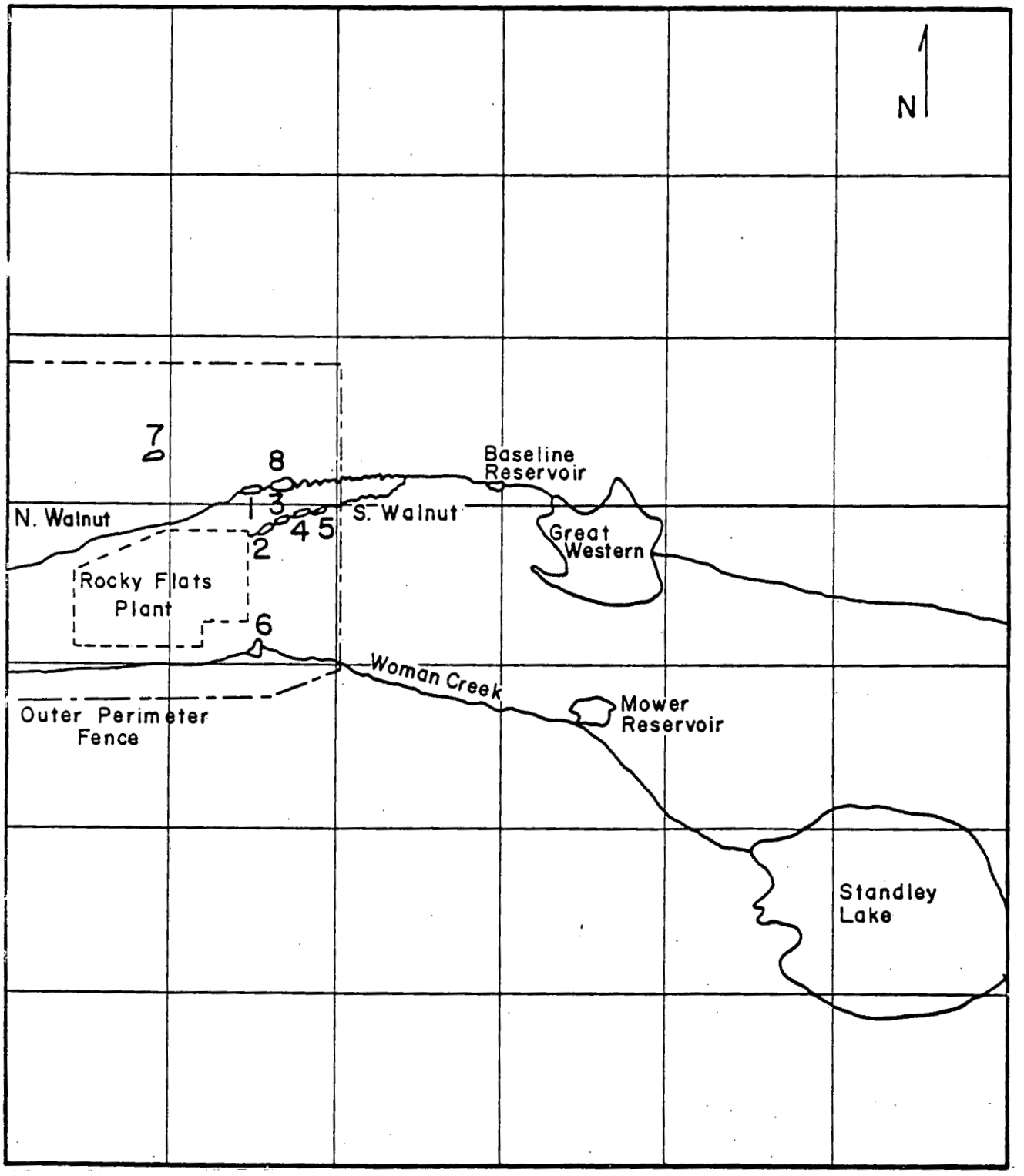

Figure 1. Study area of Rocky Flats environs showing ponds 1 - 7 and the four reservoirs. Flow on Woman and the Walnut creeks is from West to East.

II. Methods

Figure 1. gives the locations of aquatic systems in the Rocky Flats environs. Ponds 1-6 and the reservoirs are our study areas and pond 7 serves as a control pond. The new pond, designated as pond 8, will be of great interest in the future.

A. Sampling Protocol

1. Sediment

Samples of sediment are taken once per month from ponds 1 - 6 and approximately once per month from pond 7, Great Western and Standley Reservoirs. Samples of approximately 5 g of the top 3 -5 cm of the sediment ~~is~~ are collected. A grid system has been set up for each pond and sampling is uniform over each pond surface. Approximately 25 samples are taken per pond each sampling day.

2. Plants

An inventory of all major terrestrial plant species in the vicinity of ponds 1 - 6 has been completed. In addition, the species growing along the creek beds and in the vicinity of the reservoirs has been completed. Species identification were performed by Miss Sharon Svalberg. These samples were compared to those in the CSU herbarium for absolute identification (Table 1).

Aquatic plants in the water courses were also inventoried.(Table 1)

Plant samples are collected for Pu analysis at the grid markings around each pond approximately one time per month. Several of the major species are collected at each location.

Algae is collected from each pond and the reservoirs one time per month.

Phytoplankton (principally green and blue-green algae) are collected coincidentally with filtering of water samples.

Generally, the total plant sample is analyzed; however, in several instances separation of leaves, stems and fruit or flower has been performed.

3. Water

Water samples are collected at approximately 12 locations per pond once per month. One-liter samples are taken at the surface, 1/2 depth and full depth. In addition, one time per month a five-liter sample collected near the inlet and a five-liter sample collected near the outlet are composited. All water samples are filtered first through a Whatman #41 fllter. The filter residue is used for phytoplankton analysis. The water sample is then filtered through a millipore filter.

4. Animals

Animal life near the ponds and on the two creeks are collected wherever possible. Fish have been collected only in ponds 1, 6 and 7, and the reservoirs. Zooplankton is collected one time per month in ponds 1 - 6 by a nanoplankton net. Frogs, snakes and crayfish, are abundant and have been collected by trapping. To date, one doe deer from the herd that frequents the south section of the site has been obtained under a scientific collection permit.

On all mammals collected, the lung tissue, liver, bone, muscle, stomach contents and skin are analyzed. For crayfish, the exoskeleton is scrubbed to remove surface contamination.

Once a month an Ekman Dredge is used to collect a large sample of sediment from each pond. This sample is analyzed for bottom feeding organisms.

B. Analytical Protocol

The analytical method now used for plutonium analysis is a combination of the methods of Keough and Powers, and a Rocky Flats ion exchange procedure for soils.[1,2] An outline of the method is as follows:

1. Wet ash sample in Pyrex beaker with hot concentrated HNO_3 for 4 hours.
2. Ash in muffle furnace at 450°C for 7 hours.
3. Wet ash residue with hot concentrated HNO_3 for 3 hours.
4. Ash in muffle furnace at 450°C for 7 hours.
5. a) Soils - Digest in concentrated HF at low temperature in a Teflon beaker.
   b) Other samples - Digest in 6M HNO_3 + 6M HF at low temperature in Pyrex beaker.
6. Evaporate to dryness on hot plate.
7. Repeat steps 5 and 6.
8. Digest in 7.5M HNO_3 and evaporate to dryness.
9. Prepare ion exchange column.
   a) Pack column with slurry of amberlite CG-400, 100-200 mesh anion exchange resin.
   b) Pass 30 ml (10 ml at a time) 7.5M HNO_3 through column to condition.
10. Dissolve sample (step 8) in 7.5M HNO_3.
11. Allow sample to drain completely through column.
12. Rinse sample beaker 3 times with 10 ml of 7.5M HNO_3 and allow to drain through column.
13. Add 7.5M HNO_3 until effluent is colorless. (This step removes iron.)

14. Pass 20 ml. 9 M HCl through column. (Removes nitric acid from column.)
15. Add Hydroxylamine Hydrochloride crystals to top of resin bed.
16. Pass 25 ml. of 9 M HCl-5% NH_4I solution through column. Collect Pu effluent in beaker.
17. Pass 25-50 ml. of 0.5M HCl acid through column. Uranium will be present in this effluent.
18. Add 10 ml. of concentrated HNO_3 to the effluent from step 16 and evaporate to dryness on low to medium temperature.
19. Add 10 ml. of 8M HNO_3 and evaporate to dryness.
20. Dissolve in 10 ml. 2M HNO_3 -·2M Boric acid.
21. Transfer to counting vial and add 1 drop of 4M urea and 4 ml. of .6M di (2- ethythexyl) Phosphoric acid (D2EHPA) plus scintillator solution (p-terphenyl - POPOP in toluene)
22. Shake to suspend extracted Pu in organic phase.
23. Count for 100 minutes in Liquid Scintillation Counter.

Notes:

1. The recovery of the method:91 $\pm$4.6%. (1 standard deviation).
2. Counting yield on Nuclear Chicago Mark II Liquid Scintillation Counter at H-6 gain and window settings of 7%-15% is 97%.
3. Decontamination factor for natural Uranium is ~200 and for enriched Uranium ~50.
4. Minimum detectable activity (MDA) is calculated as:

MDA (95% confidence level) = $2\sqrt{\frac{2R_b}{t}}$

where R_b = background count rate

t = time

For 100 minute count the MDA is 0.18 pCi/sample.

The original Keough and Powers method is also used in many of the laboratory experiments where Uranium is not present.[1] An outline of this procedure is as follows:

1. Wet ash sample in Pyrex beaker with hot concentrated HNO_3 for 4 hours.
2. Ash in muffle furnace at 450°C for 7 hours.
3. Wet ash residue with hot concentrated HNO_3 for 3 hours.
4. Ash in muffle furnace at 450°C for 7 hours.
5. Digest in 6 M HNO_3 + 6 M HF.
6. Evaporate to dryness on hot plate.
7. Digest again in 6 M HNO_3 + 6 M HF.
8. Evaporate to dryness.
9. Digest in 8 M HNO_3 and evaporate to dryness.
10. Dissolve in hot 2 M HNO_3 + 0.2 M H_3BO_3.
11. Aliquot 10 ml. to liquid scintillation counting vial and add 1 drop 4 M urea.
12. Add 4 ml. 0.6 M di(2- ethylhexyl) phosphoric acid, (D2EHPA) + scintillator solution (p-terphenyl - POPOP in toluene).
13. Shake to suspend extracted Pu in organic phase.
14. Count for 100 min. in liquid scintillation counter.

Various solvent extraction and ion exchange procedures were investigated prior to the acceptance of the procedure now in operation. The first trial was a variation of the solvent extraction procedure by F.E. Butler[3]. An outline of the procedure is as follows:

1. Wet ash in 25 ml. hot concentrated HNO_3 and in Pyrex beaker over low to medium heat to dryness.
2. Muffle at 450°C for 3 hours.
3. Repeat steps 1 and 2.
4. Digest in 25 ml. 6 M HNO_3 - 6 M HF.
5. Evaporate to dryness.
6. Repeat steps 4 and 5.
7. Add 10 ml. of 8 N HCl to the salts twice, evaporating to dryness each time.
8. Dissolve salts in 50 ml. of 8 M HCl (with heat), and pour the solution into a separatory funnel.
9. Rinse the original container twice with 15 ml. of 8 M HCl, and add the rinses to the funnel.
10. Add 10 drops of 30% H_2O_2.
11. Add 25 ml of 10% TIOA-xylene to the funnel and shake vigorously for 10 seconds.
12. Drain and discard the aqueous layer.
13. Rinse the organic phase with 25 ml. of 8 M HCl. Discard the rinse solution.
14. Add 25 ml. of 50 to 80°C 8 M HCl -.05 M NH_4I and shake for 1 minute to strip plutonium. (Release pressure after shaking 5 seconds.)
15. Repeat step 14 and combine the two 25 ml. solutions.
16. Strip Uranium with two 25 ml. volumes of .1 M HCl and combine the solutions.

17. Evaporate each strip solution to dryness. Destroy organic residue (if any) by wet ashing with concentrated HNO_3.
18. Add 10 ml. of 8 M HNO_3 acid and evaporate to dryness.
19. Dissolve residues in 10 ml. 2 M HNO_3 -0.2M Boric acid.
20. Transfer to counting vials and add 1 drop 4 M Urea and Keough and Powers cocktail.
21. Count in liquid scintillation counter for 100 min.

No positive results were obtained using this method. Yields of Pu, enriched Uranium, and natural Uranium had large variances. The immediate cause was suspected to be the use of the 2 M HNO_3-0.2 M Boric acid in the extraction step. Therefore, the experiment was repeated as follows:

Steps 1 - 17. Unchanged

Step 18. Add 10 ml. of 8 M HCl acid and evaporate to dryness.

Step 19. Dissolve residues in 10 ml. of 3 N HCl-0.3M Boric acid.

Step 20. Transfer to counting vial and add 1 drop 4 M Urea, 5 drops .01 M Sodium dichromate, and Keough and Power's cocktail.

Again no positive results were obtained. Butler's method was abandoned because it was concluded that the Keough and Powers method using liquid scintillation counting was not compatible with the solvent extraction procedure due to factors not fully understood in environmental samples.

After negative results were obtained using Butler's procedure, a variation of the solvent extraction was attempted. The method was obtained from Rocky Flats [2]. An outline of this procedure is as follows:

1. Wet ash in 25 ml. hot concentrated HNO_3.
2. Evaporate to dryness.
3. Muffle at 450°C for 3 hours.
4. Repeat steps 1 - 3.
5. Digest in 25 ml. 6 M HNO_3 - 6 M HF.
6. Evaporate to dryness.
7. Repeat steps 5 and 6.
8. Add 10 ml. 8M HCl, evaporate at medium heat to dryness.
9. Repeat step 8.
10. Dissolve residue in 25 ml. 3 M HCl.
11. Add sample from step 10 to separatory funnel.
12. Rinse beaker with 10 ml. 3 M HCl and add this to separatory funnel in step 11.
13. Add 25 ml. 5% TIOA-xylene solution to separatory funnel.
14. Shake funnel for 10 sec.
15. Remove aqueous layer containing Pu to clean beaker.
16. Add 15 ml. of 3 M HCl-0.3 M Boric acid to separatory funnel to rinse Pu from TIOA.
17. Shake for 10 seconds and add aqueous layer to beaker in step 15.
18. Evaporate Pu solution to dryness.
19. Dissolve residue in 10 ml. 3M HCl-0.3 Boric acid and transfer to counting vial.
20. Add 20 ml. 0.1M HCl acid to separatory funnel and shake for 10 seconds.
21. Remove aqueous layer containg Uranium to clean beaker.

22. Rinse TIOA in separatory funnel with 10 ml. 0.1M HCl and add rinse to beaker in step 21.
23. Evaporate to dryness and dissolve in 10 ml. 3M HCl-0.3M Boric acid.
24. Transfer to counting vial.
25. Add 1 drop 4 M Urea and 5 drops .01 M sodium dichromate. Then add Keough and Power's cocktail.
26. Shake vials and count in liquid scintillation vial for 100 minutes.

Replicate samples of spikes solutions, both high and low activity, of ^{239}Pu, natural Uranium, and enriched Uranium resulted in highly colored solutions in the counting vials. Due to the variation in quenching, no conclusions could be drawn. At the time it was thought that perhaps there might be a $HCl-HNO_3$ complex from the initial digestion which led to the color.

Therefore, a variation of the procedure was attempted. The variations are as follows:

Step 1. Wet ash with 25 ml. hot concentrated HCl instead of HNO_3.

Step 5. Digest in 25 ml. 6 M HCl - 6 M HF instead of 6 M HNO_3 - 6 M HNO_3 - 6 M HF.

Initial experiments using spiked solutions showed promise. Recovery of ^{239}Pu was 95% and decontamination factors for Uranium and enriched Uranium were 150. However, when spiked soil samples were run through the procedure negative results again were noted. High amounts of quenching resulted. Recoveries and decontamination factors were not obtainable. Iron contamination was one reason at least for the erroneous

results. However, separation of Pu and Uranium was not efficient to any extent, leading to the conclusion that other factors in the environmental samples may have led to incomplete separation. Various scavengers were tried but were found not to be successful.

At that time it was postulated that the iron could be separated early in the procedure by ion exchange. However, the additional time and expense involved would have defeated our purposes. Therefore, solvent extraction was abandoned in favor of ion exchange.

In order to proceed with the ion exchange procedures,columns and resins had to be ordered. During this lull a preliminary experiment was done, in order to become aquainted with techniques, using materials and methods already available. The method used was a variation of that described in Lamar Johnson's thesis[4]. An outline is as follows:

1. Wet ash sample in Pyrex beaker with hot concentrated HNO_3 for 3 hours.
2. Ash in muffle furnace at 450°C for 7 hours.
3. Repeat steps 1 and 2.
4. Digest in 6 M HNO_3 - 6 M HF.
5. Evaporate ot dryness.
6. Repeat steps 4 and 5.
7. Digest in 8 M HNO_3 and evaporate to dryness.
8. Dissolve residue in 7.3 M HNO_3 and then bring to boil.
9. This solution was passed through a Dowex 1 x 8 resin column. (Dowex 1 x 2 was not available. Burettes were also used.)
10. Effluent was discarded.

11. Column was washed with 10 ml. of HCl. (Effluent contained Uranium)
12. Add Hydroxylamine Hydrochloride crystals to top of resin bed.
13. Pass a solution of .5M HCl saturated with NH_4I through column.
14. Rinse column with additional portions of .5 M HCl.
15. Combine portions 13 and 14 and wet ash with HNO_3 to destroy chloride and iodide ions.
16. Dissolve in 8 M HCl acid and evaporate to dryness.
17. Add 10 ml. of 2 M HNO_3 - .2 M Boric acid to residue.
18. Transfer to scintillation vial and add 1 drop of 4 M Urea and Keough and Power's cocktail.
19. Shake to extract Pu.
20. Count in liquid scintillation counter for 100 minutes.

Pu recoveries were from 40 - 60%. Enriched and natural Uranium contamination in the Pu effluent varied from 5% to 50%. The use of burettes and the Dowex 1 x 8 instead of Dowex 1 x 2 resin were assumed to be the factors underlying the low recoveries and high variance of Uranium contamination. Also, the Uranium might not have been thoroughly washed from the resin. Variance in flow rates using different sizes of burettes was also a factor.

After further consultation with Rocky Flats personnel the following experiment was initiated[2]:

1. Wet ash sample in pyrex beaker with hot concentrated HNO_3.
2. Ash in muffle furnace at 450°C.
3. Repeat steps 1 and 2.
4. Digest in 6 M HNO_3- 6M HF.
5. Evaporate to dryness.

6. Repeat steps 4 and 5.
7. Digest in 8 M HNO_3 and evaporate to dryness.
8. Prepare burettes.
   a) Dampen small pledget of glass wool and tamp firmly into bottom of column.
   b) Fill column with resin slurry.
   c) Condition column with 20 ml. of 7.5 M nitric acid and allow acid to pass completely through column.
   d) Repeat with an additional 10 ml. of 7.5 M HNO_3. Discard effluent.
9. Dissolve residue from step 7 in 7.5 M HNO_3.
10. Add sample solution (step 9) to column reservoir. Allow to drain completely through column.
11. Wash down the walls of the sample beaker with 10 ml. of 7.5 M HNO_3.
12. Add 5 ml. of concentrated hydrochloric acid to the column. Allow HCl to drain completely. Discard HCl effluent. Proceed immediately to following step.
13. Elute column with 10 ml. portions of .5 M HCl and allow complete drainage into a 250 ml. beaker. Save effluent.
14. Add sufficient Hydroxylamine Hydrochloride crystals to cover resin bed. Pass 10 ml. of 0.5M HCl - 10% NH_4I solution through column. Collect effluent in beaker from step 13. Repeat with additional 10 ml. of 0.5M in HCl - 10% NH_4I.
15. Add 25 ml. of concentrated HNO_3 to the beaker and evaporate to destroy solids and sublime iodine.

16. Add 8 M HNO_3 to residue and evaporate to dryness.
17. Dissolve residue in 2 M HNO_3- .2 M Boric acid and transfer to counting vial.
18. Add 1 drop of 4 M Urea and 4 ml. of Keough and Powers cocktail.
19. Shake to extract Pu.
20. Count in Liquid Scintillation Counter for 100 minutes.

Unfortunately, we discovered, after running this experiment three times, that we had been given the wrong procedure. Pu, natural Uranium and enriched Uranium, all counted in the Pu effluent. This was not designed to separate Uranium and Plutonium.

Two procedures (both quite similar) were obtained from Rocky Flats[2]. the first method was as follows:

1. Wet ash sample in Pyrex beaker with hot concentrated HNO_3 for 4 hours.
2. Ash in muffle furnace at 450°C for 7 hours.
3. Repeat steps 1 and 2.
4. Digest in 6 M HNO_3 + 6 M HF.
5. Evaporate to dryness.
6. Repeat steps 4 and 5.
7. Digest in 9 M HCl and evaporate to dryness.
8. Prepare ion exchange column until resin bed has settled. Tamp plug of glass wool firmly atop resin bed to protect it from disturbance.
9. Pass two 10 ml. portions of 9 M HCl through resin.
10. Dissolve residue in 9 M HCl and pass sample through resin bed. Discard effluent.

11. Rinse sample beaker twice with 10 ml. portions of 9 M HCl. Allow each rinse to pass completely through resin bed.
12. Pass 10 ml. portions of 8 M nitric acid through the column until yellow iron chloride is no longer visible in the effluent. Add an additional 10 ml. of 8 M nitric acid to the column. Discard all 8 M HNO_3 washings.
13. Wash excess nitric acid from the column by passing 15 to 20 ml. of 9 M HCl through column.
14. Add Hydroxylamine Hydrochloride crystals to the top of resin bed. Follow by addition of 25 ml. of 9 M HCl - 5% NH_4I solution. Collect effluent in 150 ml. beaker.
15. Add 10 ml. of concentrated HNO_3 to the eluent from step 14 and take to dryness on a low to medium temperature hot plate. Wet ash organic material remaining by repeated evaporations with concentrated nitric acid.
16. Add 10 ml. 8 M HNO_3 and evaporate to dryness.
17. Add 10 ml. 2 M HNO_3 - .2 M boric acid and transfer to counting vial.
18. Add 1 drop Urea and 4 ml. Keough and Powers' cocktail.
19. Shake to suspend Pu.
20. Count in liquid scintillation

This experiment resulted in very low recoveries which were highly inconsistant for ^{239}Pu. Poor separation of Uranium and Plutonium also resulted. Since the second procedure was run concurrently with this procedure and excellent results were obtained, no further time has as yet been spent in order to answer the questions as to why this failed.

The second procedure is the Plutonium analysis procedure mentioned at the beginning of this section and is now being used to analyze Rocky Flats environmental samples.

A variation of this procedure is being investigated where a solvent extraction step (TIOA) is inserted after the ion exchange step. This will enable, hopefully, higher discrimination factors for enriched and natural Uranium when very low level samples are involved.

Most of the previously mentioned procedures were found not to be compatible when combined with HF digestion and liquid scintillation counting techniques. Because of limited time and a desire to obtain a workable procedure that was satisfactory, many of the reasons as to why the methods failed were not investigated. However, it should be noted that most analytical procedures for Pu determination are not applicable to environmental samples. Most are designed for laboratory experiments where there is no interference from other radionuclides. Many of the procedures for environmental samples are either time consuming and expensive, or are designed for mg. quantities of Pu. A multitude of interferences and inconsistancies seem to appear when µg. quantities are investigated. In order to do a proper environmental sampling program, with µg. quantities of Pu and other radionuclides which interfere, many samples must be taken in order to insure good statistical results. A procedure must be relatively inexpensive and simple enough for any technician to follow, unless of course, a large laboratory is available which has an unlimited budget and many employees.

Much more work needs to be done concerning analytical procedures and their practical application to long range environmental sampling.

References

1. Keough, R.F. and G.J. Powers, Analytical Chemistry, 42:419, 1970.
2. Rocky Flats consultations.
3. Butler, F.E., "Rapid Bioassay Methods for Plutonium, Neptunium and Uranium." Health Physics, 15:19-24, 1968
4. Lamar Johnson, C.S.U. Thesis, 1969.

TABLE 1. PLANTS OF THE ROCKY FLATS WATER COURSES

Plant	Common Name	Period of Active Growth
Family:Alismaceae		
Sagittaria cuneata	Sagittaria	July - August
Family:Asclepiadaceae		
Asclepias speciosa	Showey Milkweed	July - October
Asclepias stenophylla	Narrowleafed Milkweed	July - October
Family:Boraginaceae		
Mertensia lanceolata	Chiming Bells	April-early June
Family:Cactaceae		
Echinocactus simpsonii	Mountain Ball Cactus	perennial- blooms in April
*Opuntia polycantha	Plaines Prickley Pear	perennial- blooms in July
Opuntia rafinesquei	Prickley Pear Cactus	perennial-blooms in July
Family:Caprifoliaceae		
Symphoricarpos occidentalis	Snowberry bush	Perennial- blooms in late June
Family:Caryophyllaceae		
Cerastium arvense	Mouse ear	late April - early June
Family:Chenopodiaceae		
*Chenopodium leptophyllum	Goose foot	June - late August
*Kochia iranica	Burning bush	April-September
*Salsola kali	Russian Thistle (Tumbleweed)	May-September
Family:Commelinaceae		
Tradescantia occidentalis	Spiderwort	April- June
Family:Compositeae		
Achillea lanceolata	Yarrow	Late June-Sept.
*Ambrosia spp.	Ragweed	June-August
Artemesia frigida	Fringe Sage	July-October
Cirsium arvense	Canadian Thistle	May-September
Erigeron speciousus	Fleabane	May-June
*Gaillardia aristata	Blanket Flower	late June - August
Grindelia squarrosa	Gum weed	late July - October
Gutierrezia sarothrae	Turpentine weed	August-October

TABLE 1. (Continued)

Plant	Common Name	Period of Active Growth
Helianthus annuus	Sunflower	July-October
*Lactuca scariola	Wild Lettuce	June-September
Liatris punctata	Dotted Gay feather	August-October
Ratibida columnifera	Cone flower	August-September
Senecio atratus	Butter weed	September - October
*Stephanomeria pauciflora	Wire Lettuce	April-September
Taraxacum officinale	Dandelion	late March - early November
Tragopogon dubius	Goats beard	April - June
Xanthium strumarium	Cockleburr	late July - October
Family:Convulvulaceae		
Convulvulus arvensis	Bindweed	May-July
Family:Crucifereae		
Descurainia sophia	Tansey Mustard	late March - early November
Erysimum asperum	Wallflower	late March - July
Lepidium campestre	Pepper grass	June-September
Rorippa islandica	Cress	June-September
Thlaspi alprestre	Pennycress	March-July
Family:Cyperaceae		
Carex filifolia	Sedge	May-September
Cyperus filiculmis	Flatsedge	June-September
Scirpus microcarpus	Bulrush	July-August
Family:Euphorbiaceae		
Euphorbia dictyosperma	Spruge	July-September
Euphorbia marginata	Snow-on-the-mountain	July-September
Family:Geraniaceae		
Erodium cicutarium	Heron bill	late March-September
Geranium fremontii	Cranes bill	May-September
Family:Gramineae		
Agropyron smithii	Western wheatgrass	June-September
Andropogon gerardii	Big bluestem	June-October
Bromus inermis	Sleepy grass	April-October
Setaria viridis	Green bristle grass	April-October
Sitanion hystrix	Squirrel tail	May-August
Stipa comata	Needle and thread	May-July
Stipa neomexicana	New Mexico feather grass	May-July

TABLE 1. (Continued)

Plant	Common Name	Period of Acitve Growth
Family:Hypericaceae		
Hypericum perforatum	St. Johnswort, Klamath weed	June-early October
Family:Iridaceae		
Sisyrinchium montanum	Blue-eyed grass	May-July
Family:Juncaceae		
Juncus balitcus	Rush	April-October
Family:Labitae		
Mentha arvensis	Mint	June-early October
Monarda fistulosa menthaefolia	Bee balm	July-August
Scutellaria brittonii	Skull cap	late April-June
Family:Leguminoseae		
Amorpha nana	False indigo bush	April-October blooms in June
Astragalus spp.	Milk vetch	April-July
Glycyrrhiza lepidota	Wild licorice	June-September
Lathyrus eucosmus	Peavine	May-July
Melilotus alba	White sweet clover	May-August
Melilotus officinalis	Yellow sweet clover	April-August
Oxytropis spp.	Loco-weed	June-August
Petalostemum purpurea	Prarie clover	July-August
Thermopsis divaricarpa	Golden banner	May-early July
Family:Linaceae		
Linum lewisii	Flax	May-June
Family:Malvaceae		
Sphaeralcea coccinea	Globe mallow	June-July
Family:Najadaceae		
Potamogeton natans	Pondweed	June-September
Family:Onagraceae		
Gaura coccinea	Butterfly weed	May-September
Oenothera brachycarpa	Evening primrose	May-July
Family:Papaveraceae		
Argemone polyanthemos	Prickley poppy	May-July
Family:Polygonaceae		
Rumex crispus	Dock	April-October
Family:Ranunculaceae		
Ranunculus aquatilis	Water crowfoot	May-July

TABLE 1. (Continued)

Plant	Common Name	
Ranunculus glaberrimus	Buttercup	April-July
Family:Rosaceae		
Prunus virginiana	Choke cherry	April-September-blooms in May
Rosa woodsii	Woods rose (bush)	April-September.
Family:Scrophulariaceae		
Linaria dalmatica	Toadflax	April-October
Mimulus floribundus	Monkey flower	June-August
Penstemon angustifolius	Penstemon	May-July
Scrophularia lanceolata	Figwort	May-July
Verbascum thapsus	Mullein	April-October
Veronica americana	Speedwell	May-October
Family:Solanaceae		
Solanum elaeagnifolium	Silverleaf nightshade	May-October
Solanum rostratum	Buffalo burr	June-August
Family:Typhaceae		
Typha latifolia	Cattail	April-October-blooms in July
Family:Violaceae		
Viola nuttallii	Wild violet	April-June

TABLE 2. ANIMALS OF THE ROCKY FLATS WATER COURSES

Class - Amphibia
- *Rana pipiens* — Leopard Frog

Class - Aves
- *Agelaius phoeniceus* — Red-winged Blackbird
- *Anas platyrhynchos* — Mallard
- *Charadrius vociferus* — Killdeer
- *Sturnella magna* — Meadowlark
- *Zenaidura macroura* — Dove

Class - Mammalia

Order - Artiodactyla

Family:Cervidae
- *Odocoileus hemionus* — Mule deer

Order - Carnivora

Family: Canidae
- *Canis latrans* — Coyote

Family:Mustelidae
- *Mephitis mephitis* — Striped Skunk
- *Taxidea taxus* — American Badger

Family:Procyonidae
- *Procyon lotor* — Raccoon

Order - Lagomorpha

Family:Leporidae
- *Sylvilagus audubonii baileya* — Cottontail Rabbit

Order - Rodentia

Family:Cricetinae
- *Microtus pennsylvanicus modestus* — Meadow Mouse (prarie vole)
- *Ondatia zibethicus cinnamomina* — Muskrat
- *Peromyscus maniculatus osgoodi* — White-footed Deer Mouse
- *Peromyscus nasustus* — Long-nosed Deer Mouse

Family:Geomidae
- *Thomomys talpoides* — Northern pocket gopher

Family:Sciuridae
- *Spermophilus tridecemlineatus* — 13 Lined Ground Squirrel

Class - Reptilia

Order - Chelonia
- *Chrysemyap bellii* — Painted Box Turtle

Order - Squamata
- *Crotalus viridis viridis* — Prairie Rattle Snake

TABLE 2. (Continued)

Pituophis catenifer sayi	Bull Snake (gopher snake)
Thamnophis radix	Plains Garter Snake

TABLE 3. Aquatic life of the Rocky Flats Water Courses

Cladocera	Aquatic Insects
Daphnia pulex	Plecoptera (Stoneflies)
Copepoda	Ephemeroptera (Mayflies)
Diaptomus	Odonata (Damsel & Dragon Flies)
Malacostraca	Diptera (Flies)
Gammarus	Coleoptera (Beetles)
Decapoda	Trichoptera (Caddisflies)
Crayfish *Cambarus*	

TABLE 4 Algae Found in the Rocky Flats Water Courses

CYANOPHYTA Blue-Green	CHLOROPHYTA Green	CHRYSOPHYTA
Gleotrichia	Hydrodictyon	Dinobryon
Gleocapsa	Chlorococcum	Cymbella
Oscillatoria limnosa (Roth)	Chlorella	Hylotheca
Nostoc pruniforme	Oedogonium	Navicula
Anabaena	Cladophora	
Scytonema	Zygnema	Desmids
Stigonema	Ulothrix zonata	Closterium
Tolypothrix	Chaetophora	
Aphanizonema	Pediastrum	Euglenophyta
Amphithrin	Stigeoclonium	Euglena
Calothrix	Spirogyra crassi	
	Spirogyra	
	Euglenoids	
	Scenedesmis	
	Mougeotia	

B. Linnological Data from the Rocky Flats Water Courses

Because of thick ice over the ponds, malfunctions in our hydrolab equipment and the construction, we have limnological data for only three months. We hope to have a more thorough sampling program next year.

TABLE 5. Limnological Data From the Rocky Flats Water Courses

Pond 1

Date	Depth	Water Temperature °C	Dissolved O_2 (ppm)	Conductivity (μmho/cm)	pH	Air Temp. °C
April 4, 1972	Surface	10	10.8	0.150	7.3	15
	0.5 Meters	10	10.8	0.150		
	1.0 M	10	11.0	0.160		
	1.5 M	10	11.0	0.150		
	2.0 M	9	11.3	0.160		
	2.5 M	9	11.4	0.160		
May 10, 1972	Surface	12	7.80	0.110	7.1	10
	0.5 M	12	7.90	0.110		
	1.0 M	12	7.85	0.115		
	1.5 M	11.5	7.75	0.120		
	2.0 M	11	7.70	0.120		
	2.5 M	10	7.65	0.110		
June 30, 1972	Surface	22	8.9	0.190	7.7	27
	0.5 M	22	8.7	0.200		
	1.0 M	22	8.6	0.200		
	1.5 M	21.5	8.6	0.200		
	2.0 M	20	5.9	0.200		
	2.5 M	20	5.6	0.210		

TABLE 5. Limnological Data (Continued)
Pond 2

Date	Depth	Water Temperature °C	Dissolved O_2 (ppm)	Conductivity (μmho/cm)	pH	Air Temp. °C
April 4, 1972	Surface	13	8.15	0.205	7.3	15
	0.5 M	12.5	7.6	0.205		
	1.0 M	11.5	6.0	0.190		
	1.5	11.5	6.2	0.200		
May 10, 1972	Surface	15	6.25	0.260	7.7	9
	0.5 M	15	6.40	0.260		
	1.0 M	15	5.30	0.280		
June 30, 1972	Surface	23	5.3	0.34	7.6	
	0.5 M	21	5.0	0.340		
	1.0 M	20.5	4.8	0.330		

TABLE 5. Limnological Data (Continued)
Pond 3

Date	Depth	Water Temperature °C	Dissolved O_2 (ppm)	Conductivity (µmho/cm)	pH	Air Temp. °C
April 4, 1972	Surface	11	9.5	0.200	8.3	13
	0.5 M	11	8.5	0.205		
	1.0	10	8.7	0.210		
	1.5 M	10	8.6	0.210		
May 10, 1972	Surface	14	89.5	0.200	8.0	11.5
	0.5 M	14	8.20	0.210		
	1.0 M	15	8.10	0.210		
	1.5 M	13	8.00	0.220		
June 30, 1972	Surface	23.5	9.0	0.380	7.9	27.5
	0.5 M	23	8.8	0.380		
	1.0 M	22.5	8.8	0.380		
	1.5 M	21.5	8.9	0.390		

TABLE 5. Limnological Data (Continued)
Pond 4

Date	Depth	Water Temperature °C	Dissolved O_2 (ppm)	Conductivity (μmho/cm	pH	Air Temp. °C
April 4, 1972	Surface	11	8.9	0.200	7.4	12.5
	0.5 M	10	8.0	0.200		
	1.0 M	10	7.8	0.200		
	1.5 M	10	8.2	0.210		
	2.0 M	10	8.3	0.220		
May 10, 1972	Surface	13	8.5	0.240	7.0	12
	0.5 M	13	8.8	0.235		
	1.0 M	13.5	8.8	0.235		
	1.5 M	14	8.8	0.235		
	2.0 M	14	7.6	0.230		
June 30, 1972	Surface	24	1.59	0.200	6.8	26
	0.5 M	24	8.7	0.190		
	1.0 M	22.5	7.8	0.190		
	1.5 M	22.5	6.7	0.190		
	2.0 M	22	5.0	0.200		

TABLE 5. Limnological Data (Continued)
Pond 5

Date	Depth	Water Temperature	Dissolved O_2 (ppm)	Conductivity (µmho/cm)	pH	Air Temp. °C
April 4, 1972	Surface	10	11.3	0.08	8.2	14
	0.5 M	10	10	0.085		
	1.0 M	10	10.5	0.10		
	1.5 M	10	10.5	0.10		
	2.0 M	10	---	0.10		
May 10, 1972	Surface	14	11.2	0.100	8.5	10
	0.5 M	14	11.2	0.120		
	1.0 M	14	11.6	0.135		
	1.5 M	14	11.4	0.140		
	2.0 M	14	11.4	0.150		
	2.5 M	12	4.4	0.150		
June 30, 1972	Surface	23	14.3	0.110	8.0	26
	0.5 M	23	13.0	0.110		
	1.0 M	23	9.7	0.120		
	1.5 M	22.5	8.8	0.140		
	2.0 M	22	7.6	0.140		

TABLE 5. Limnological Data (Continued)
Pond 6

Date	Depth	Water Temperature °C	Dissolved O_2 (ppm)	Conductivity (μmho/cm)	pH	Air Temp. °C
April 4, 1972	Surface	9	12.8	0.17	6.7	16.5
	0.5 M	9	12.6	0.17		
	1.0 M	9	13.0	0.17		
May 10, 1972	Surface	12.5	10.8	0.190	6.9	12
	0.5 M	12.5	10.8	0.190		
	1.0 M	12.5	10.9	0.190		
	1.5 M	12.5	10.9	0.190		
June 29, 1972	Surface	20	9	0.090	7.3	27
	0.5 M	17.5	8	0.090		
	1.0 M	16	8.5	0.090		
	1.5 M	16	8			

C. Model of Rocky Flats Aquatic System

Figure 2. represents the probable interrelations between inorganic substances, plants and animals in the Rocky Flats environs. Arrows indicate the path of materials from the primary (inorganic) sources through various organisms and back to the soil, water, or air. Any one circuit is a food chain. All food chains in a community constitute a food web. Food webs are very complex, but may be illustrated by two simplified examples:

1) Aquatic

Bacteria and Diatoms → Small Protozoans → Larger Protozoans → Rotifers and Small Crustaceans → Aquatic Insects → Fishes → Large Carnivores and Man.

2) Land

Plants → Insects, Rodents and Grazing Mammals → Predacious Insects and Small Carnivores → Large Carnivores and Man.

Each food web at any intermediate or final step, ends in death and decay, becoming food for bacteria thus completing the circuit. The food in any community is characterized by a pyramid of numbers. Organisms at the bottom are small and abundant, whereas those at the top are few but large. The food web is highly affected by external stimuli. This can be readily observed in the Rocky Flats environs.

The biomass (total amount of living material in a given area or the total of all members of a species in an area) at Rocky Flats is in constant turmoil. The introduction of inorganic and organic wastes,

construction, and high winds have resulted in striking effects. The most obvious effects resulting from the remodeling of the holding ponds are discussed elsewhere in this report. The food webs of the seven ponds currently under study vary with location and the above mentioned effects. Nutrients, light, temperature, oxygen, siltation and wastes are the primary contributors to this variation.

Pond 7 represents the ideal situation. Nutrients are readily available, oxygen is high, siltation is at a minimum, and wastes from Rocky Flats are not present. Light easily penetrates to the bottom resulting in an abundance of higher plant life. Fish, turtles, frogs, phytoplankton, zooplankton and aquatic insects are plentiful. Everything exists as part of a well balanced, healthy system.

Ponds 1 and 6 exhibit a similar type of balance, though less developed. There are fewer species and numbers of higher aquatic plants, fewer bottom organisms, and smaller fish. All of these are probably related to siltation and nutrient availability.

Ponds 2-5 show a marked decrease in biomass due to high siltation, introduction of treated sewage, increased temperatures and little light penetration resulting in odiferous muck rich in hydrogen sulfide and methane due to larger amounts of organic material reaching the bottom than can possibly be utilized by bottom fauna. In fact, little, if any bottom fauna are found in these ponds, which is mute testimony that highly toxic substances detrimental to aquatic life are present. The characteristic brown color of the ponds is still further evidence. Zooplankton populations are extremely low, and the phytoplankton that does exist is characteristic of polluted waters.

Although this may be one factor, many other factors are involved and simple aeration will not solve the problem.

Aeration of the ponds will lead to an increase in suspended particulates resulting in a detrimental effect upon fish and will also prevent new particles from settling out. This could easily lead to a higher output of plutonium from the ponds.

In order for fish to survive in the holding ponds, thermopollution, siltation, industrial wastes, and nutrient releases would have to be better controlled. This would help alleviate the detrimental effects upon fish and lower organisms leading to a normal healthy food chain.

References

1. Smith, L., Ecology and Field Biology, Harper and Row, 1966.
2. Storer, T.I. and R.L. Usinger, General Zoology, 4th Edition. McGraw-Hill Book Co., 1965.

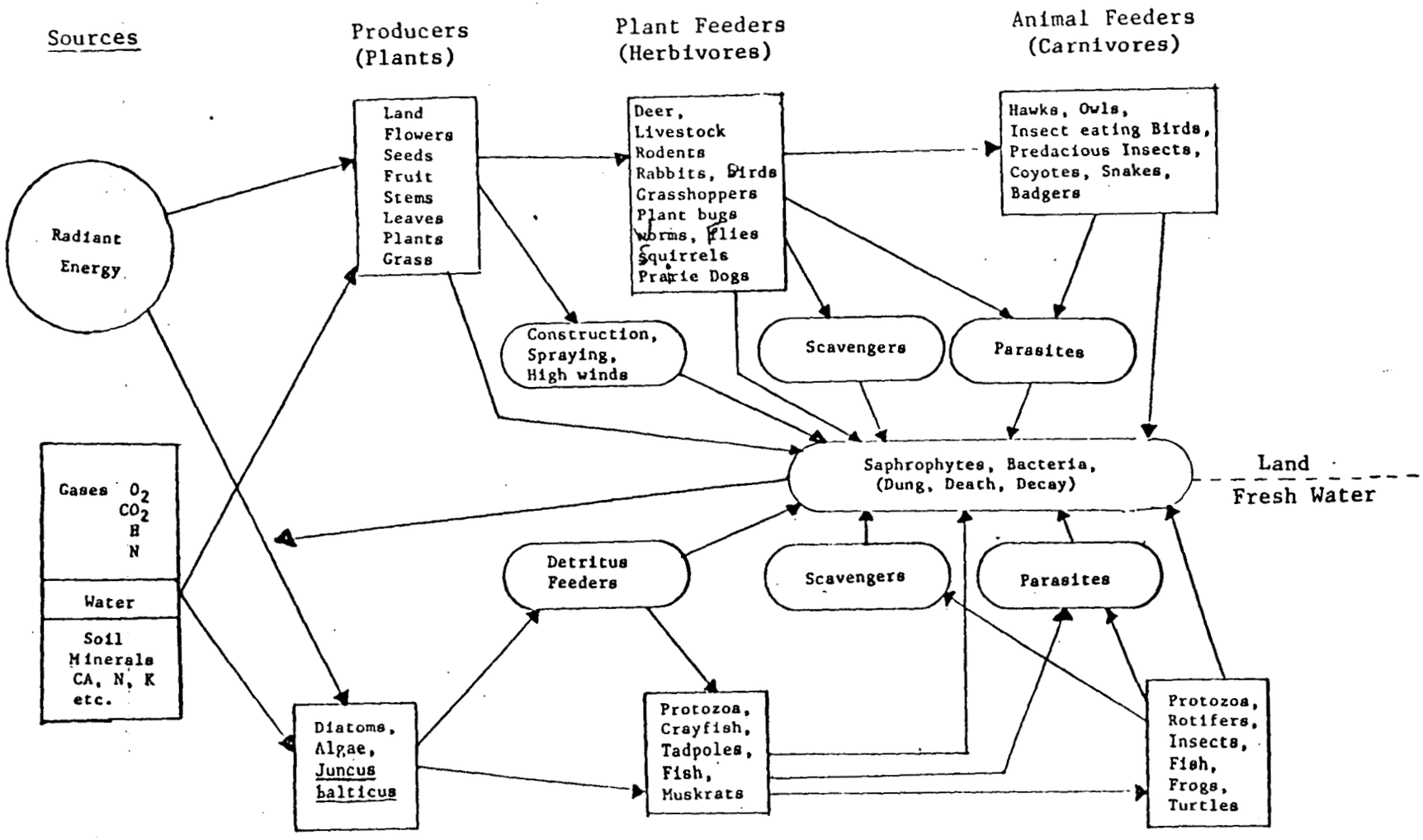

Figure 2. Model of Rocky Flats Aquatic System.

D. Description of Effects of Pond Reconstruction Activity

Because of the contruction around the ponds, we have not been able to continue our plant and animal inventory to any great extent. However, we have added a few more genera to the lists. These plants are either early spring bloomers or late summer bloomers that we had not identified by report time last year. Any new plant entries next year will be meaningless in terms of the plant commumities of the sites when the project began. This is because the process of succession has been set back by the construction and the area will again have to pass through the preliminary successional stages to eventually approach an ecosystem comparable to that of a year ago.

This spring before construction began, the progression of plant succession was apparent, The frequency of weedy annuals had decreased while native biannuals and perennials (grasses mostly) increased. This was a small step toward the climatic grassland ecosystem of the area.

The extensive devastation of the landscape due to the construction work has caused severe disruption of the soil integrity. The topsoil has been scraped away leaving immature undeveloped soils, deficient in organic matter, one of the most important constituents of any soil. Besides keeping the soil loose and friable, it greatly increases the water holding capacity. Organic matter is the source of three essential elements; nitrogen, phosphorus and sulfur. It also chelates iron, zinc, and copper in soluble forms, making them available for plant utilization. Without sufficient organic matter in the soil, plants will show a wide range of deficiency symptoms which will persist until the nutrient balance has once again been established.

Next spring the construction scars will be partially obscured by the invasion of weedy species that can tolerate these low nutrient levels, especially low nitrogen levels. Most of the leguminous plants are a good example of these. Among the first species to emerge will be those with highly developed wind dispersal mechanisms such as Tragopogon, Taraxacum , Typha, Cersium, Aristida, and Asclepias. Then will come the gradual intrusion of and domination by the grasses of the surrounding undistrubed areas. Within a decade the vegetation around the ponds should approach what it was a year ago, but will never again attain the exact same community composition.

Before we can continue the extensive sampling we have done in the past it will be necessary to resurvey all of the ponds. We will redetermine sediment volume as well as water volume. The surface areas and the perimeters of the ponds have been changed by the construction work, as have the flow patterns of the water. It will take a few months for the sediment accumulations to equilibrate with the new flow patterns of the water. This is the only major long-term effect of the construction on the ponds themselves. Because of the undeveloped states of the ponds and because their bottoms were not dredged, they should recover from this construction within a year or so.

References

1. Smith, L., Ecology and Field Biology, Harper and Row, 1966.
2. Storer, T.I. and R.L. Usinger, General Zoology, 4th Edition. McGraw-Hill Book Co., 1965.

E. Plutonium Concentrations in Components of the Aquatic System

A great percentage of effort during the second year was devoted to improving analytical techniques and quantifying results of environmental sampling with laboratory experimentation. Therefore, only a relatively few new environmental samples have been fully analyzed. Approximately 800 new samples and approximately 700 samples undergoing reanalysis with new techniques are now at various steps of completion. A few of the initial reanalyzed sample results are shown in Table 6. These initial results show that previously analyzed samples are not in serious error. Further analysis of all reanalyzed samples will hopefully confirm these initial findings. The fact that Uranium quenches at a much higher degree than Plutonium may be the reason for the lack of a large error in the samples analyzed the first year.

TABLE 6. Reanalysis of Previous Samples

Sample #	Type Sample	1971 pCi/gm	1972 pCi/gm	%
61	14 - 3 sediment	88.8	71.7	80.7
76	14 - 18 sediment	61.2	34.5	56.4
83	13 - 7 sediment	92.6	85.7	92.5
86	13 - 10 sediment	105	91.1	86.7
91	13 - 15 sediment	426	386	86.5
115	12 - 1 sediment	178	171	96
129	12 - 9 sediment	502	475	94.6
134	12 - 14 sediment	232	166	71.6
239	22 - 1 water	21.9	19.8pCi/1	90.5
272	24 - 10 water	4.4	3.16pCi/1	71.9
464	33 plant	49.1	48.3	98.5
539	46 animal	16.0	17.0	106

$\overline{X}$ = 85.99 %

F. Summary of Results of Other Investigations

It is valuable to compare the results of other investigations of Pu contamination in our study area.

The Colorado Department of Health[1] measures Pu concentration in Walnut Creek at Indiana monthly. They reported an average monthly value of 2.85 pCi/liter for 1971. For 1970 their average value was 0.88 pCi/liter. This can be compared to a value of 0.13 pCi/liter reported by Dow Rocky Flats for 1969. Comparing gross alpha activity for the same periods at the same site, the Department of Health reported an average value of 17 pCi/liter, while Dow reported 2.2. It appears that the discrepancy is due to the method of analysis. The Health Department does not filter the water before analysis and as a result it must contain appreciable seston , i.e. plant and animal life which possess high concentration factors for Pu and U. Poet and Martell[3] reported values of 0.2 and 0.82 pCi/liter for two samples in the summers of 1969 and 1970, respectively. They did not give their sample handling procedure. The EPA reported a value of 0.05 pCi/liter for water at the same site in February, 1970. In June of 1971, we observed 0.85 pCi/liter for a 5 liter sample from the same site, however, this was for an unfiltered water sample.

Sediment sample data may also be compared. Poet and Martell report an average value of 4.5 pCi/g for the pond at Walnut Creek in Indiana in 1969. Our values averaged 1.3 pCi/gram for 1971.

For Great Western Reservoir, Poet and Martell found 0.065 and 0.21 pCi/gram for two samples in 1969.and 1970. Dow reported an average value of 0.45 for 9 samples in 1969. The EPA reported an average value of 0.11 pCi/gram. Our values for June of 1971 were less

than 0.05 pCi/gram. There are no vegetation or animal data to directly compare with ours.

References

1. U.S. AEC Rocky Flats Plant, 1971, Environmental Surveillance Summary. Report, Colorado Department of Health.

2. Radiation Data and Reports, 13:584-587, 1972.

3. S.E. Poet and E.A. Martell, Health Physics, 23:537-548, 1972.

4. Radioactivity in the Environs of the Rocky Flats Plutonium Plant, EPA, Water Quality Office, 1971.

IV. Laboratory Experiments on Plutonium Uptake

1. Microbial Uptake of Plutonium

The possibility that bacteria in the aquatic system might take up Pu was recognized early. Workers have shown that bacteria take up and immobilize appreciable lead in the cell wall and membrane fraction.[1] To investigate if this occurred for Pu we performed the following experiments:

1. Micrococcus luteus and Pseudomonas aeruginosa, two common bacteria, were grown in a solution of $^{239}Pu(NO_3)_4$ and nutrient media. The preparations were incubated at 25°C with agitation in a rotary shaker. The cells were harvested by centrifugation and washed sucessively with water, Ivory soap flake solution and DTPA. The percentage of plutonium in each wash and in the cells is shown in Table 7.

It is clear from ~~this~~ these data that there was appreciable uptake of Pu by the bacterial cells. There was greater uptake per gram of Pseudomonas cells, a gram-negative bacterium. The uptake was 12 times greater by the Pseudomonas. In the lead up take experiments[1], the gram-negative bacteria took up 30 times as much as the Micrococcus, a gram-positive bacterium. Experiments are planned for this year to determine if the Pu is associated with the cell wall and membrane fraction or with the cytoplasmic fraction.

2. The above experiment was repeated using high fired PuO_2, the only difference being that the PuO_2 was contained in a dialysis bag. In this experiment the average uptake of Pseudomonas was 300% per gram of bacterial cells and for Micrococcus 14% per gram of cells. This is the same order as in the previous experiment, Pseudomonas exhibiting 21 times greater uptake. However, the results of this experiment are

TABLE 7. Microbial Uptake of $^{239}Pu(NO_3)_4$

A. Micrococcus luteus

	Water Wash %Pu	Ivory Soap Wash %Pu	DTPA Wash %Pu
Medium	98.7	84.6	90.3
Wash 1	5.2	7.4	5.7
Wash 2	1.2	1.8	1.7
Wash 3	0.6	2.8	1.1
Bacterial cells	3.2	3.5	1.8

Average uptake per gram of Bacteria = 2.33%/gram

B. Pseudomonas aeruginosa

	Water Wash %Pu	Ivory Soap Wash %Pu	DTPA Wash %Pu
Medium	75.6	70.8	73.8
Wash 1	5.7	10.6	10.3
Wash 2	5.0	7.0	3.9
Wash 3	1.4	1.8	1.2
Bacterial cells	12.3	9.8	10.7

Average uptake per gram of Bacteria - 27.3%/gram

intriguing. For reasons unknown at this time the uptake per gram was nearly 10 times greater for both bacterial types. This is interesting because the Pu was contained in a dialysis bag and even if the smaller or soluble particles escaped the bag, there is no reason to suspect the uptake should be greater than for the $^{-}NO_3$ form. Experiments are planned to answer this phenomenon.

References

1. T.G. Tornabene and H.W. Edwards. Microbial uptake of Lead. Science, 176:1334-1335, 1972.

2. Uptake of Plutonium by Plants and Plant Roots

The main consideration in any food chain ultimately leading to man is biological uptake of ^{239}Pu by primary producers (plants). The controversy arises when considering whether the plutonium contamination is actually uptake or merely surface contamination. E.M. Romney, and others[2] have indicated that relatively small amounts of ^{239}Pu are transferred from soil to plants through roots. Concentration factors of 10^{-4} to 10^{-5} were noted. Their conclusions assume that the high concentration factors from environmental samples are due almost entirely to external surface contamination. In order to explore this controversy, the following preliminary experiments were performed.

Barley plants were grown in nutrient media from seeds in order to test the active transport of plutonium by plant roots. Roots were cut from the barley plants and enclosed in cheesecloth packets. They were then introduced into a solution containing soluble $^{239}Pu(NO_3)_4$. At certain intervals a packet of roots was removed and washed with distilled water in order to remove the majority of the surface contamination. The roots were then analyzed for ^{239}Pu. A tentative concentration factor from solution to roots of approximately 200 was observed. This is evidence that root uptake is not selective against $^{239}Pu(NO_3)_4$. With such a large concentration factor in roots, an experiment was designed to investigate the uptake into the aerial portion of the plant.

Barley plants were again grown from seeds in greenhouse conditions until root length was sufficient. The roots were then introduced into solutions of complete media spiked with $^{239}Pu(NO_3)_4$ or high fired

$^{239}PuO_2$. The complete media contained all nutrients required for healthy plant growth. It should be noted that at no time did the aerial portion of the plant come into direct contact with the spiked solutions. Concentration factors from the roots to the aerial portion are given in Table 8. These concentration factors are in the same ranges as those given by Romney for soil to aerial plant portions. No significant difference was noted between the $^{239}PuO_2$ and the $^{239}Pu(NO_3)_4$ uptake, and an increase in uptake over time was apparent for both.

These experiments show that at least in a non-stressed environment rich in nutrients, there does appear to be a selective mechanism which discriminates against plutonium reaching the aerial portion of the plant.

In conclusion, roots appear to concentrate ^{239}Pu in relatively large amounts, but transfer of ^{239}Pu to the rest of the plant is negligible, at least in non-stressed conditions. There is evidence that this is not the case under stressed conditions. ^{210}Po, for instance, will accumulate to a higher degree when plants are in a stressed "hunger state". More investigations need to be conducted concerning uptake in stressed versus non-stressed conditions, but at this point, we can say that surface adsorption appears to be the main source of contamination in environmental plant samples.

References

1. E.M. Romney, H.M. Mork and K.H. Larson, "Persistance of Plutonium in Soil, Plants and Small Mammals". Health Physics, Pergamon Press, 19:487-491 (1970).

2. D.O. Wilson and J.F. Cline, Nature, Lond. 209, 941 (1966).

3. G.E. Powers. (Personal Communication)

TABLE 8. Uptake of Pu by Plants

Sample	Time of Harvest (Hrs.)	Sample Type	Activity/g	Concentration Factor
Control 1		Total	1.10 pCi	
Control		Roots	2.90	
PuO_2 - 1	1059	Total	4.70 pCi	
PuO_2 - 1		Roots	1130 pCi	$4x10^{-3}$
PuO_2 - 2	1059	Total	1.60 pCi	
PuO_2 - 2		Roots	1180 pCi	$1x10^{-3}$
PuO_2 - 3	1059	Total	2.90 pCi	
PuO_2 - 3		Roots	3400 pCi	$8x10^{-4}$
$PuNO_3$ - A	138	Total	72.6 pCi	
$PuNO_3$ - A		Roots	145,000 pCi	$5x10^{-4}$
$PuNO_3$ - 1	138	Total	7.4 pCi	
$PuNO_3$ - 1		Roots	6460 pCi	$1.1x10^{-3}$
$PuNO_3$ - 2	48	Total	0.388 pCi	
$PuNO_3$ - 2		Roots	170,000 pCi	$2.2x10^{-6}$
$PuNO_3$ - 3	138	Total	210 pCi	
$PuNO_3$ - 3		Roots	125,000 pCi	$1.6x10^{-3}$
$PuNO_3$ - 4	138	Total	35.9 pCi	
$PuNO_3$ - 4		Roots	141,000 pCi	$2x10^{-4}$
$PuNO_3$ - 5	72	Total	103 pCi	
$PuNO_3$ - 5		Roots	107,000 pCi	$9x10^{-4}$
$PuNO_3$ - 6	138	Total	36.5 pCi	
$PuNO_3$ - 6		Roots	132,000 pCi	$2x10^{-4}$

3. Uptake of Plutonium by Freshwater Fish

Because fish are a possible link to man in an aquatic food chain, the possibility of their concentrating ^{239}Pu is of importance. A preliminary experiment was designed to examine the transfer of ^{239}Pu from water to fish. Goldfish were introduced into an aquarium containing 22 liters of water spiked with $^{239}Pu(NO_3)_4$ and neutralized to a pH of 7.4. Because a charcoal filter could not be used in the aquarium, the fish survived in this confined environment for only 96 hours. Fish and water samples were taken periodically. The results are in Table 9 and Figure 3. In order to insure no surface contamination or cross contamination from dissection on the relatively small fish, the outer epidermal and scaley portion of the fish was sloughed off by immersing the fish in a beaker of concentrated nitric acid. This left only fish muscle and bone to be analyzed. A concentration factor of 100 was noted after 96 hours.

Little data is available from the literature for concentration factors from fresh water in fish. However, Adams and Fowler[1] show a concentration factor from water to fish using $^{238}PuO_2$ microspheres of approximately 1000. Pillai[2] showed relatively low concentration factors for marine fish. More intricately designed experiments over a longer time period are planned in order to better determine the concentration factors in fish. These will be done to investigate not only transfer from water but also transfer to fish through the food chain which should be of more importance since fish ingest very little water.

References

1. W.H. Adams and Eric B. Fowler, "^{238}Pu incorporated in fish living in water containing $^{238}PuO_2$".

2. D.C. Pillai, Dr. R.C. Smith, Dr. T.R. Folsom, "Plutonium In the Marine Environment". Nature, 203:568-571, Aug. 8, 1964.

TABLE 9. Accumulation of ^{239}Pu by Goldfish

Sample	Time (4 hrs.)	Fish cpm/gm.	Water cpm/ml
0	.00	--	48
1	.08	24	41
2	.17	45	55
3	.50	308	51
4	2.00	769	41
5	3.50	654	47
6	5.50	1308	49
7	21.30	1081	44
8	93.50	2780	31
9	96.50	2787	28
10	98.00	1210	21

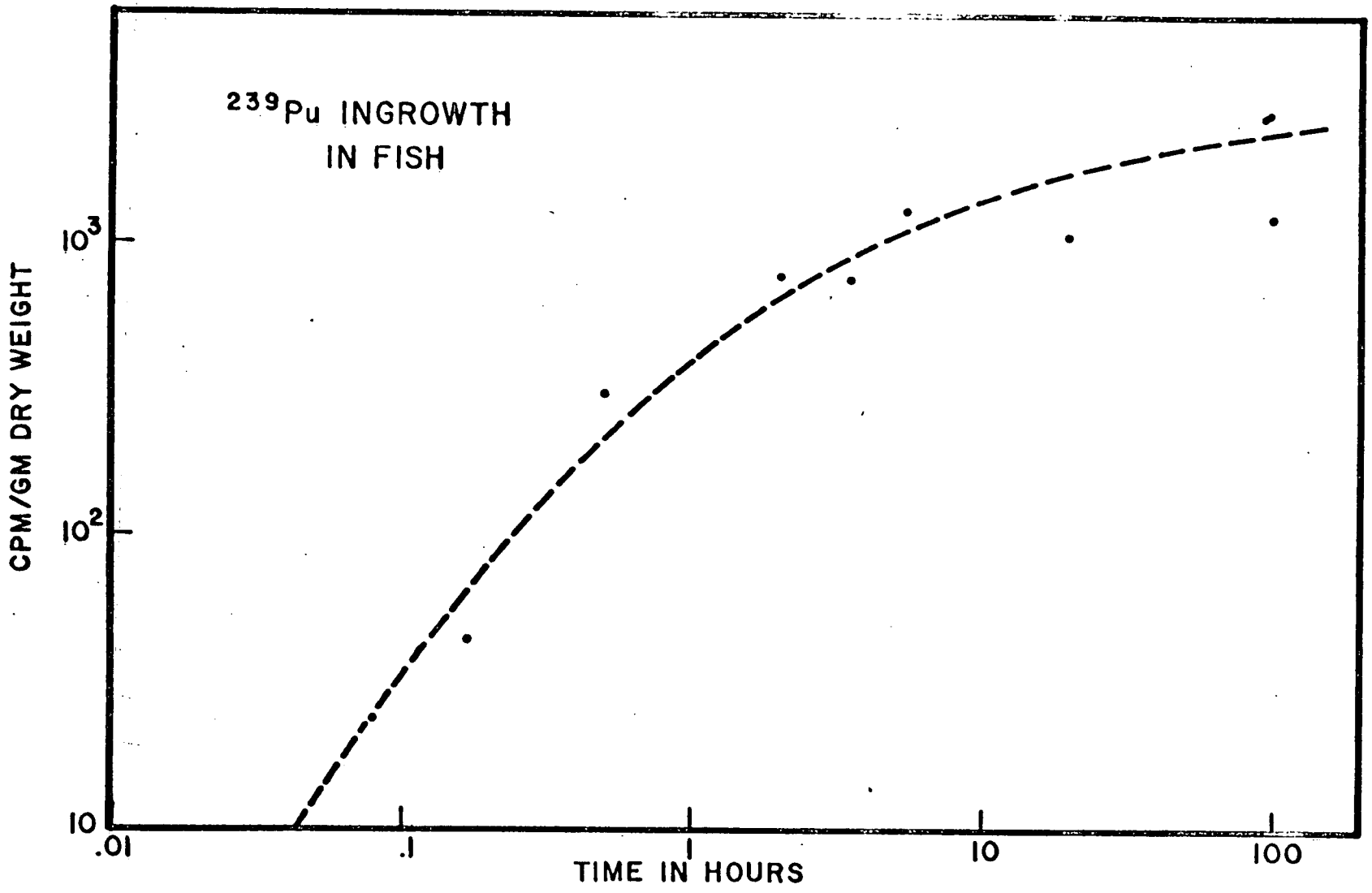
239Pu INGROWTH
IN FISH
CPM/GM DRY WEIGHT
10³
10²
10
.01
.1
1
10
100
TIME IN HOURS

Figure 3.

4. Transfer Kinetics of Pu from Water to Sediment

Experiments were performed studying the transfer of Pu from water to sediment. Rocky Flats pond water was spiked with $Pu(NO_3)_4$ and allowed to contact sediment from Pond 7. The disappearance of activity from the water was studied as a function of time. The function appeared to consist of two exponential terms and was described by the equation:

$$C(t) = C_o(0.75\ e^{-0.723t} + 0.25\ e^{-0.066t})$$

where: $C(t)$ is the concentration of Pu in water at any time t

t = time in days

$C_o(t)$ = initial concentration of Pu in water.

This experimental finding fits remarkably well with actual pond limnological data. The average Walnut Creek flow into Pond 2 during 1971 was measured to be 480 M^3/day. The volume of Pond 2 was calculated to be 1500 M^3. Therefore, the mean lifetime of any parcel of water in Pond 2, if mixing is uniform, can be calculated as follows:

$$\bar{t} = \frac{1500\ M^3}{480\ M^3/day} = 3.1 \text{ days}$$ (3½ days)

From the equation above, 78% of the Pu in water delivered to Pond 2, on the average, should be exchanged to sediment in an average residence time of 3.1 days.

From average sediment Pu concentrations determined in 1971 and measurement of sediment depths it was estimated that the inventory of Pu in the 4 holding ponds on Walnut Creek was as follows:

Pond 2	1.75×10^5 pCi
Pond 3	0.23×10^5 pCi
Pond 4	0.23×10^5 pCi
Pond 5	0.05×10^5 pCi

From this it can be calculated that Pond 2 contains approximately 77% of the Pu in the total of the four. It must be fortuitous that the numbers are so close, as often the water flow is shunted by some of the ponds. Also, the data for Pond 4 does not agree with what would be postulated using the above simple approach.

It is however, encouraging and additional experiments will be performed to describe the retention function more accurately and see what parameters affect it, e.g. pH, Pu chemical form, temperature.

5. Transfer of Pu from Water to Algae

Algae constitute by far the majority of the aquatic plant material found in the holding pond chain on Walnut Creek. In the previous year of study, we observed that the transfer from water to algae was extremely high. In fact, much higher than previously observed for marine systems [1, 2]. This should be explained by the fact that the mineral content of ocean water is so much greater than fresh water that the turnover rate must be correspondingly greater. Thus, the equilibrium concentrations in marine algae would be proportionately less and the concentration factors less.

The concentration factors we reported in last years report were in the order of 10^4 to 5×10^4.

An experiment was designed to study the uptake of Pu by algae under controlled conditions. We used two types of algae. Chlorella, a unicellular spherical algae and Spirogyra a filamentous, colonial type. Of the two chlorella has the greatest surface area. We spiked the nutrient media containing the algae with either $^{239}Pu(NO_3)_4$ or $^{239}PuO_2$. The concentration factors observed after 1 week are shown in Table 10.

The data from this preliminary experiment suggest that uptake by algae is more than simply surface adsorption. This is concluded from the observation that the uptake seemed independent of the surface area of the algae type. Also, the uptake was significantly greater for the more soluble $^{-}NO_3$ form.

TABLE 10. Uptake of Pu by Algae

Algae	Pu Chemical Form	Mean Concentration Factor
Chlorella	$^{239}Pu(NO_3)_4$	40,600
	$^{232}PuO_2$	9,800
Spirogyra	$^{239}Pu(NO_3)_4$ *	42,000
	$^{239}PuO_2$	6,800

* C.F. after 2 days due to necrosis.

References

1. G.G. Polikarpov, in: "Radioecological Concentration Processes" (B. Aberg and F.P. Hungate, editors) pp. 819-825, Pergamow, New York (1967).

2. Victor E. Noshkin, Health Physics, 22, 537 (1972)

V. Conclusions

After two full years of field and laboratory study on the movement of Pu in a fresh water system, we still can make only a few tentative conclusions. This is because (1) a great percentage of effort during this second year was devoted to improving our analysis technique and (2) our sampling protocol was seriously disturbed by pond construction activities.

It does appear however, that:

(3) The Pu concentrations reported in the previous annual report[1] are not in serious error.

(4) Bacteria do take up significant Pu activity, but it is not clear whether this Pu is more or less mobile in any natural food chain.

(5) The uptake by fresh water algae is extremely high and dependent upon the chemical form of the Pu.

(6) The drastic physical changes in the existing ponds and the construction of a new pond will allow observation of the kinetics of Pu buildup in various conponents and species.

(7) The exchange of Pu from water to sediment is very rapid. The kinetics of exchange observed in laboratory experiments appear to describe the concentrations found in the Rocky Flats holding pond system.

References

1. J.E. Johnson, The Study of Plutonium in Aquatic Systems of the Rocky Flats Environs. First Technical Progress Report, Colorado State University, 1971.

VI. Publications

D. Paine, J.E. Johnson, and R.L. Watters. "Plutonium Movement in Aquatic Systems: A Review.", Proceedings of the Rocky Flats Symposium on Safety in Plutonium Handling Facilities, CONF-710401, United States Atomic Energy Commission, 1971.